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## **Frequency Upshifting of Laser Pulses by Ionization Fronts**

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# FREQUENCY UPSHIFTING OF LASER PULSES BY IONIZATION FRONTS

The propagation of electromagnetic radiation through plasmas is a problem of general interest with a wide variety of applications ranging from communications<sup>1</sup> to laser driven particle accelerators.<sup>2,3</sup> For example, recent plasma simulation studies<sup>4,5</sup> suggest two possible methods by which the frequency of an electromagnetic (EM) wave may be upshifted. In the first method,<sup>4</sup> the plasma density through which the EM wave is propagating is suddenly increased in time, while the second method<sup>5</sup> utilizes the interaction of a plasma wave wakefield (having a phase velocity near the speed of light)<sup>2,3,6,7</sup> with a short EM pulse.<sup>8</sup> In this Letter, a novel method<sup>9</sup> for upshifting the frequency of a laser pulse is proposed and analyzed, which utilizes the interaction of a laser pulse with a relativistic ionization front. In such a scheme, an intense driving electron beam (or, alternatively, an ionizing laser beam) is used to generate an ionization front, propagating near the speed of light, with the plasma density rising from zero to some high value  $n_0$  over a relatively short distance  $L$ . A "test" laser pulse of length  $< L$  which is properly phased, such that it co-propagates with the ionization front in the region of the large negative local density gradient, will be continuously frequency upshifted as it propagates. This mechanism may provide a practical and efficient method for tuning the frequency of a laser pulse, in which the upshift is adjusted by varying the ionization density and/or the interaction distance.

The physical mechanism by which an ionization front may be used to upshift the frequency of a laser pulse may be understood by the following. Consider a laser pulse of frequency  $\omega$  and wavenumber  $k$ , co-propagating in the region of the ionization front, both of which are assumed to be moving at approximately the speed of light. Furthermore, assume that in the region of the laser pulse, the ionization front may be approximated by a linear density gradient of the form

$$n \simeq \begin{cases} \bar{n}, & \xi < -L; \\ -\bar{n}\xi/L, & -L < \xi < 0; \\ 0, & \xi > 0; \end{cases} \quad (1)$$

where  $\xi = z - ct$  and  $L$  is the length of the ionization front (see Fig. 1). For the present discussion,  $\bar{n} = n_0$  and  $L = L_0$  are assumed to be constant. Consider a laser pulse centered at  $\xi_c = -L_0/2$ , with a pulse length of  $\ell$ , where  $L_0 > \ell \gg \lambda = 2\pi/k$ , and of initial frequency  $\omega_0 \gg \omega_{pe} = (4\pi e^2 n_0 / m_e)^{1/2}$ . If the frequency shift induced in the pulse

remains small compared to  $\omega_0$ , the local phase velocity of the pulse (via the dispersion relation  $\omega^2 = \omega_p^2 + c^2 k^2$ , where  $\omega_p^2 = 4\pi e^2 n/m_e$ ) is approximately  $v_p/c \simeq 1 + \omega_{p0}^2 n/2\omega_0^2 n_0$ . Local variations in the plasma density  $n(\xi)$  lead to variations in the local phase velocity  $v_p(\xi)$  of the laser pulse. For example, the local phase velocity near the front of the laser pulse  $v_p(\xi_+)$  will be less than that near the back of the pulse  $v_p(\xi_-)$  provided  $n(\xi_+) < n(\xi_-)$ . Hence, the individual phase peaks in the pulse  $\sim \exp(ikz - i\omega t)$  may move relative to one another (i.e., closer together for the present example). The resulting change in the radiation wavelength may be estimated by  $\lambda(\tau) \simeq \lambda_0 + \Delta v_p \tau$ , where  $\Delta v_p \simeq \lambda_0 dv_p/d\xi$  is the difference in phase velocity between adjacent phase peaks,  $\lambda_0 \simeq 2\pi c/\omega_0$  is the initial wavelength and  $c\tau$  is the laser/ionization front propagation distance. This gives  $\lambda/\lambda_0 \simeq 1 - c\tau\omega_{p0}^2/2L_0\omega_0^2$ , which corresponds to an increase in frequency of  $\omega/\omega_0 \simeq 1 + c\tau\omega_{p0}^2/2L_0\omega_0^2$ . By a similar mechanism, a plasma wave may be used to induce shifts in the frequency of a laser pulse.<sup>5,8,9</sup>

One expects the above estimates to hold in the limit in which the frequency shifts remain small,  $\Delta\omega = \omega - \omega_0 \ll \omega_0$ . However, it is possible to analytically obtain expressions for large frequency shifts ( $\Delta\omega \geq \omega$ ) in the limit  $\omega_p^2/\omega^2 \ll 1$  via the wave equation. The transverse plasma response current, in the fluid approximation, is  $J_\perp \simeq -en v_\perp$ , where  $v_\perp$  is the transverse electron fluid velocity. In 1D, conservation of canonical transverse momentum implies  $v_\perp \simeq ca$ , which is the quiver motion of the electrons in the laser field assuming  $|a|^2 \ll 1$ . Here,  $a = eA_\perp/m_e c^2$  is the normalized vector potential of the laser. Using the variables  $\xi = z - ct$  and  $\tau = t$ , the transverse wave equation is given by

$$\left( \frac{2}{c} \frac{\partial}{\partial \xi} - \frac{1}{c^2} \frac{\partial}{\partial \tau} \right) \frac{\partial}{\partial \tau} a = \frac{\omega_{p0}^2}{c^2} \frac{n(\xi, \tau)}{n_0} a(\xi, \tau), \quad (2)$$

where  $n(\xi, \tau)$  is the plasma electron density of the ionization front which is assumed to be known and independent of the vector potential of the short laser pulse.

The wave equation may be solved analytically for the case of a square laser pulse of the form  $a = \hat{a} \exp(i\omega\xi/c)$ , for  $-\ell < \xi < 0$ , and  $a = 0$ , otherwise. Also, a linear ionization density profile will be assumed, of the form given by Eq. (1). In general, the pulse amplitude  $\hat{a}$ , frequency  $\omega$ , pulse length  $\ell$ , ionization density  $\bar{n}$  and ionization front

length  $L$  are all functions of  $\tau$ . In the limit  $\omega_p^2/\omega^2 \ll 1$ , the second order  $\tau$  derivative in Eq. (2) may be neglected, and one finds  $\hat{a} = a_0\omega_0/\omega$  for  $-\ell < \xi < 0$ , where

$$\frac{\omega(\tau)}{\omega_0} = \left[ 1 + \frac{\omega_{p0}^2}{\omega_0^2} \int_0^\tau d\tau' \frac{c\bar{n}(\tau')}{n_0 L(\tau')} \right]^{1/2}, \quad (3)$$

with  $n_0 = \bar{n}(\tau = 0)$ ,  $a_0 = \hat{a}(\tau = 0)$  and  $\omega_0 = \omega(\tau = 0)$ . Notice that as the laser pulse evolves, the laser power  $P \sim |\partial a / \partial \xi|^2$  is approximately constant in  $\tau$ , i.e., frequency upshifts correspond to decreases in the laser amplitude,  $\hat{a}(\tau) \sim 1/\omega(\tau)$ .

By integrating the wave equation in  $\xi$  over the longitudinal extent of the laser pulse, an equation for the total normalized pulse energy  $U \equiv \int d\xi |\partial a / \partial \xi|^2$  may be derived. In the limit  $\omega_p^2/\omega^2 \ll 1$ , and for the linear density gradient given by Eq. (1), the pulse energy evolves according to  $\partial U / \partial \tau = \alpha \omega_{p0}^2 \bar{n} / 2n_0 c L$ , where  $\alpha = \int d\xi |a|^2 > 0$ . Hence, the laser pulse gains energy from the ionization front as it propagates. For the case of a square pulse of length  $\ell$ , one finds  $\ell(\tau)/\ell_0 = \omega(\tau)/\omega_0$ , where  $\ell_0 = \ell(\tau = 0)$ , which implies that the pulse length increases as the frequency is upshifted.

For a “fixed”, externally generated ionization front (i.e., generated by an ionizing pump laser or electron beam) in which  $\bar{n} = n_0$  and  $L = L_0$  are independent of  $\tau$ , Eq. (3) gives

$$\omega(\tau) = \omega_0 \left( 1 + c\tau \omega_{p0}^2 / L_0 \omega_0^2 \right)^{1/2}. \quad (4)$$

Equation (4) indicates that asymptotically, for  $c\tau \gg L_0 \omega_0^2 / \omega_{p0}^2$ , the frequency scales as  $\omega \simeq \omega_{p0} (c\tau / L_0)^{1/2}$ , where  $c\tau$  represents the distance the pulse has propagated through the plasma. Notice that frequency shifts  $\Delta\omega \sim \omega_0$  require  $c\tau \simeq L_0 \omega_0^2 / \omega_{p0}^2$ . In principle, Eq. (4) indicates that there is no upper limit to how far the frequency may be upshifted (assuming that the ionization front and laser pulse may be sustained over a sufficiently large distance within the plasma), however, this is a result of assuming  $v_0 \simeq v_g \simeq c$ , where  $v_0$  is the velocity of the ionization front and  $v_g$  is the group velocity of the laser pulse. In practice,  $v_0 \neq v_g$  which implies that the laser pulse will “phase slip” in  $\xi$  out of the region of the ionization front ( $-L_0 < \xi < 0$ ) and, thus, will no longer be frequency upshifted. Consider a laser pulse with a pulse center  $\xi_c$  initially located at  $\xi_c(\tau = 0) = \xi_{c0}$ , where

$-L_0 < \xi_{c0} < 0$ . Phase slippage will cause  $\xi_c$  to evolve in  $\tau$  according to  $\partial\xi_c/\partial\tau = v_g - v_0$ . Assuming  $v_0 \simeq c$ ,  $v_g/c \simeq 1 - \omega_{p0}^2 n(\xi_c)/2\omega^2 n_0$  and  $n(\xi_c) = -n_0 \xi_c/L_0$ , where  $\omega = \omega(\tau)$  is given by Eq. (4), gives  $\xi_c(\tau)/\xi_{c0} = \omega(\tau)/\omega_0$ . The detuning distance  $c\tau_d$ , defined to be the propagation distance required for the laser pulse to phase slip out of the ionization front,  $\xi_c(\tau_d) = -L_0$ , is given by  $c\tau_d = L_0(\epsilon^{-2} - 1)\omega_0^2/\omega_{p0}^2$ , where  $\epsilon \equiv |\xi_{c0}/L_0|$ . Inserting this into Eq. (4) gives a maximum frequency upshift of  $\omega(\tau_d) = \omega_0/\epsilon$ . For example, a pulse initially centered at  $\xi_{c0} = -L_0/4$  ( $\epsilon = 1/4$ ) gives a final frequency of  $\omega(\tau_d) = 4\omega_0$  after a detuning distance of  $c\tau_d = 15L_0\omega_0^2/\omega_{p0}^2$ .

The above analysis has assumed that i) the velocity of the ionization front is constant,  $v_0 \simeq c$ , as may be the case for an ionization front produced by a relativistic electron beam, and ii) the density profile of the ionization front  $n(\xi)$  was assumed independent of the propagation time  $\tau$ . The limit placed on the frequency shift as a result of phase slippage may be circumvented by relaxing either of these two conditions.

If the ionization front is directly produced by the laser pulse itself, then the condition  $v_0(\tau) = v_g(\tau)$  may automatically be satisfied. When  $v_0 = v_g$ , there is no slippage and, hence, the frequency of an intense laser pulse may be "self-upshifted" as a result of the interaction of the pulse with its self-generated ionization front. For a square pulse which generates an ionization front with a uniform gradient  $dn/d\xi = -n_0/L_0$ , the frequency evolves according to Eq. (4). Since  $v_0 = v_g$ , the absence of slippage implies that the upshifting process will be limited by some other mechanism, such as laser pulse diffraction. As an example, consider a 100 fs ( $\ell_0 \simeq 30 \mu\text{m}$ ), KrF laser pulse ( $\lambda_0 = 2\pi c/\omega_0 \simeq 0.25 \mu\text{m}$ ) ionizing hydrogen gas at 1 atm ( $\lambda_{p0} = 2\pi c/\omega_{p0} \simeq 4.5 \mu\text{m}$ ). In order to double the laser frequency, an interaction distance of  $c\tau \simeq 3.0 \text{ cm}$  is required. Assuming that the laser pulse propagation distance is limited by diffraction implies  $c\tau \simeq 2Z_R$ , where  $Z_R = \pi r_s^2/\lambda_0$  is the vacuum Rayleigh length and  $r_s$  is the laser spot size. Setting  $2Z_R \simeq 3.0 \text{ cm}$  gives  $r_s \simeq 34 \mu\text{m}$ . The energy required to ionize this volume of hydrogen at 1 atm is about 11 mJ. Hence, a laser pulse energy on the order of 100 mJ should be sufficient.<sup>10</sup> This process of frequency self-upshifting may be of practical significance, however, a detailed analysis of this process requires the dynamics of laser induced ionization be included self-consistently.

Alternatively, the limitations resulting from phase slippage, which occur for the case of an externally generated ionization front, may be eliminated by gradually increasing the plasma density of the ionization front as a function of the propagation time  $\tau$ . By correctly tailoring the ionization density as a function of  $\tau$ , it may be possible to control the laser group velocity such that  $v_0 = v_g$ , thus eliminating phase slippage. The requirement that  $v_g = v_0$  is constant independent of  $\tau$ , implies that density must be tailored such that  $n(\tau) \sim \omega^2(\tau)$ , i.e.,  $L_0 \bar{n}(\tau)/L(\tau)n_0 = \omega^2/\omega_0^2$ . Solving for the ratio  $R = \bar{n}(\tau)/L(\tau)$  gives  $R/R_0 = \exp(c\tau\omega_{p0}^2/L_0\omega_0^2)$ , where  $R_0 = R(\tau = 0)$ . Hence, the ionization profile must be tailored such that  $\bar{n}/L$  is an exponentially increasing function of  $\tau$ , which may be achieved by using additional lasers to control the ionization rate. For such a density profile, Eq. (3) indicates that the frequency is exponentially upshifted

$$\omega(\tau) = \omega_0 \exp(c\tau\omega_{p0}^2/2L_0\omega_0^2). \quad (5)$$

Zero slippage further requires that the ionization front velocity be held constant at  $v_0/c = 1 - |\xi_{c0}|\omega_{p0}^2/2L_0\omega_0^2$ , which may be satisfied by adjusting the energy of the driving electron beam.

To validate the analytical results, the full wave equation, Eq. (2), is solved numerically for a laser pulse having a half-sine initial profile,  $|a| = |\sin(\pi\xi/\ell_0)|$  for  $-\ell_0 < \xi < 0$ , as shown in Fig. 1(a), for the parameters  $\lambda_0 = 2.5 \mu\text{m}$ ,  $\lambda_{p0} = 10 \mu\text{m}$ ,  $\ell_0 = 25 \mu\text{m}$ ,  $L_0 = 50 \mu\text{m}$  and  $v_0 = c$ . Equation (2) is solved for two cases: (i)  $\bar{n} = n_0$  and  $L = L_0$  are constant, which is assumed in deriving Eq. (4), and (ii)  $L = L_0$  is constant and  $\bar{n} = n_0 \exp(c\tau\omega_{p0}^2/2L_0\omega_0^2)$ , which is assumed in deriving Eq. (5). The results, after propagating a distance  $c\tau = (2\ln 2)L_0\lambda_{p0}^2/\lambda_0^2 \simeq 0.11 \text{ cm}$ , are shown in Fig. 1(b) for case (i) and in Fig. 1(c) for case (ii). The results show excellent agreement with the analytic predictions (which assumed a square pulse) to within 5%. Specifically, Fig. 1(b) indicates that  $\omega \simeq 1.5\omega_0$  ( $\lambda \simeq 0.65\lambda_0$ ),  $|a| \simeq 0.65|a_0|$  and  $\ell \simeq 1.5\ell_0$ , in agreement with the analytic theory,  $a \sim 1/\omega$  and  $\ell \sim \omega$ , where  $\omega$  is given by Eq. (4). Likewise, Fig. 1(c) indicates that  $\omega \simeq 2\omega_0$  ( $\lambda \simeq 0.5\lambda_0$ ),  $|a| \simeq 0.5|a_0|$  and  $\ell \simeq 2\ell_0$ , in agreement with the analytic theory where  $\omega$  is given by Eq. (5). The profile distortion and the “tail” observed behind the pulse in Figs. 1(b) and 1(c) may be attributed to longitudinal dispersion arising from the initial half-sine profile.



The above analysis indicates that the use of a relativistic ionization front to upshift the frequency of a laser pulse may be of practical significance. Physically, frequency upshifts arise from the pulse propagating in the region of the ionization front in which the local density gradient is negative. This produces a negative gradient in the local phase velocity within the pulse, which allows the laser wavelength to decrease and the frequency to increase as the pulse propagates. Interaction with an ionization front having a uniform gradient,  $dn/d\xi = -n_0/L_0$ , evolves the frequency as described by Eq. (4),  $\omega \sim \tau^{1/2}$ . The laser pulse evolves such that the power is constant,  $|a| \sim 1/\omega$ , whereas the pulse energy increases due to increases in the pulse length,  $\ell \sim \omega$ . Furthermore, phase slippage limitations may be overcome by appropriately increasing the plasma density as a function of propagation time  $\tau$  which may lead to substantially higher frequencies, i.e., as described by Eq. (5). Hence, ionization fronts, by varying the ionization density and/or the interaction length, may provide a convenient method for continuously upshifting laser pulse frequencies. In addition to this mechanism, an ionization front may also be used as a relativistic plasma mirror upon which may be reflected an oppositely directed laser pulse.<sup>9,11-13</sup> The reflected radiation will be upshifted by the relativistic doppler factor,  $\omega_s \simeq 4\gamma_0^2\omega_i$ , where  $\omega_i$  is the frequency of the incident radiation,  $\omega_s$  is the frequency of the scattered (reflected) radiation and  $\gamma_0^2 = 1/(1 - v_0^2/c^2)$  is the relativistic factor associated with the ionization front. This process is currently being investigated and will be the subject of future publication.

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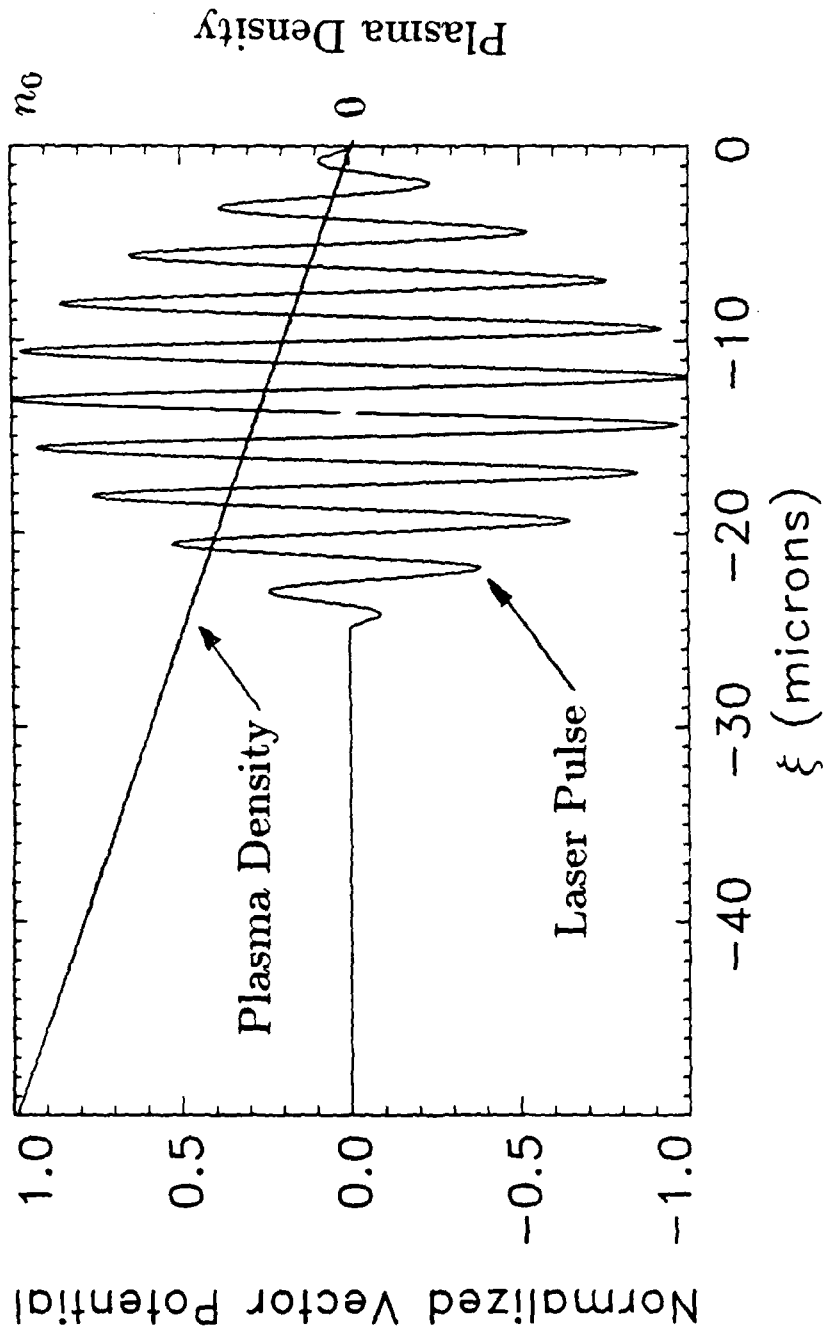
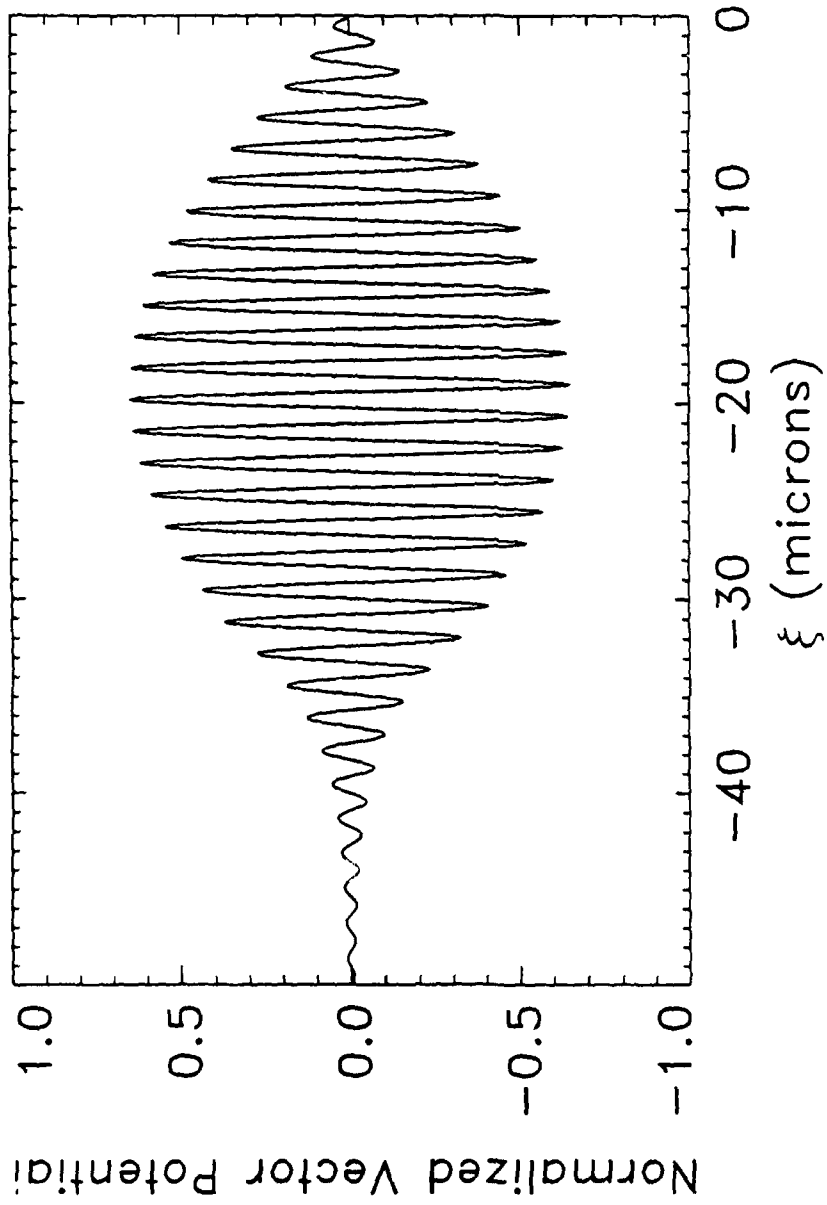
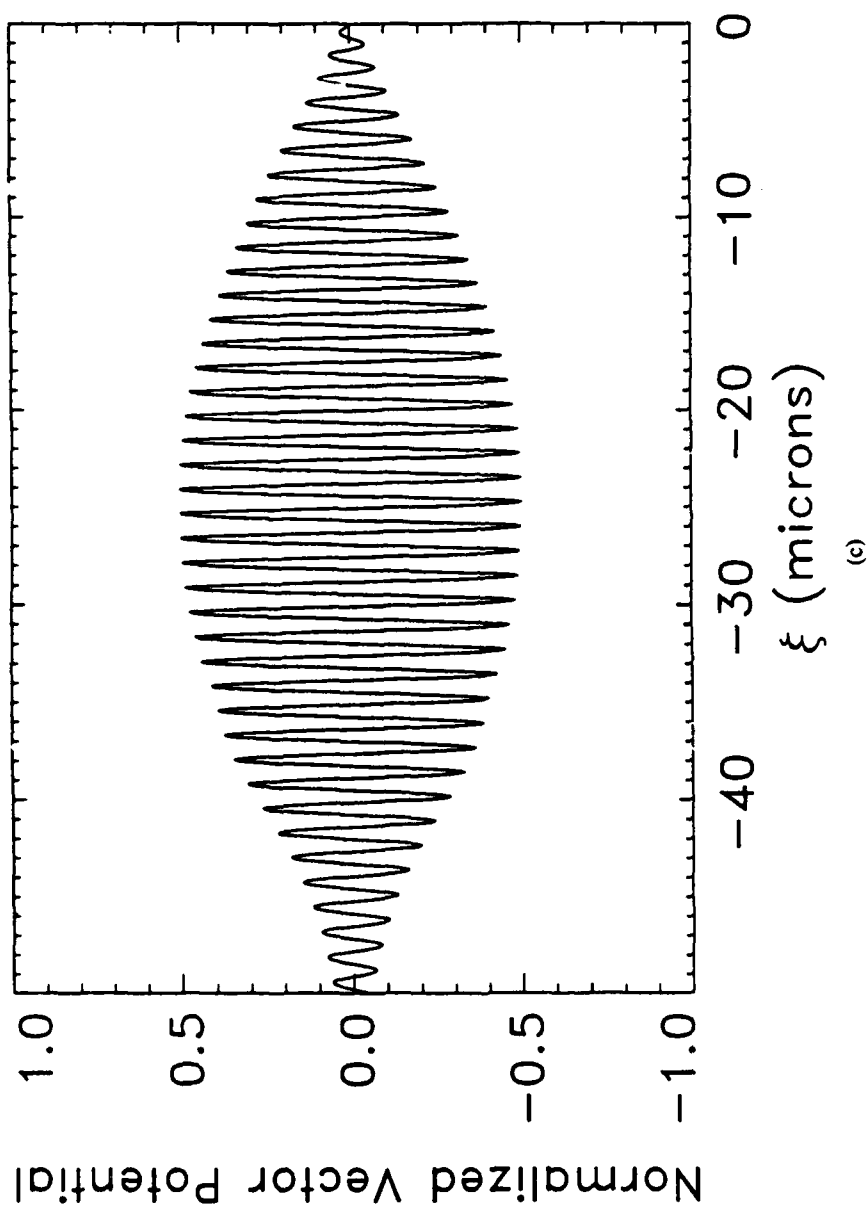


Fig. 1 — The initial laser pulse and ionization density are shown in (a) as a function of  $\xi = z - ct$ . Numerical solutions of laser pulse evolution after propagating 0.11 cm are shown in (b) for  $L$  and  $\bar{n}$  constant, and in (c) for  $L$  constant and  $\bar{n}$  an increasing function of  $\tau$ .



(b)

Fig. 1 — (Continued) The initial laser pulse and ionization density are shown in (a) as a function of  $\xi = z - ct$ . Numerical solutions of laser pulse evolution after propagating 0.11 cm are shown in (b) for  $L$  and  $\bar{n}$  constant, and in (c) for  $L$  constant and  $\bar{n}$  an increasing function of  $\tau$ .



(c)

Fig. 1 — (Continued) The initial laser pulse and ionization density are shown in (a) as a function of  $\xi = z - ct$ . Numerical solutions of laser pulse evolution after propagating 0.11 cm are shown in (b) for  $L$  and  $\bar{n}$  constant, and in (c) for  $L$  constant and  $\bar{n}$  an increasing function of  $\tau$ .